PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference AJL/CT/P5049P5093	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/GB 03/02761	International filing date *(day/month/year)* 26.06.2003	Priority date *(day/month/year)* 17.07.2002
International Patent Classification (IPC) or both national classification and IPC G01F23/26, G01F23/26		
Applicant EPICHEM LIMITED et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 4 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 8 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the opinion
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
22.01.2004	14.06.2004
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Fenzl, B Telephone No. +49 89 2399-2783

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB 03/02761

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, Pages

1, 6-11	as originally filed
2-5, 5a	received on 13.05.2004 with letter of 10.05.2004

Claims, Numbers

1-22	received on 13.05.2004 with letter of 10.05.2004

Drawings, Sheets

1/5-5/5	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).

   ☐ the language of publication of the international application (under Rule 48.3(b)).

   ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   ☐ contained in the international application in written form.

   ☐ filed together with the international application in computer readable form.

   ☐ furnished subsequently to this Authority in written form.

   ☐ furnished subsequently to this Authority in computer readable form.

   ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   ☐ the description, pages:

   ☐ the claims, Nos.:

   ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-22
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-22
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-22
	No:	Claims	

2. Citations and explanations

   see separate sheet

The invention relates to an apparatus for monitoring the level of a liquid in a vessel (claim 1), a bubbler containing an organometallic compound (claim 21) and a method for monitoring the level of an organometallic compound (claim 22).

Problem: continuously monitoring the level of a reactive liquid in a vessel.

Solution: using a metallic probe hermetically sealed within the vessel, the sealing end of the probe being encased within a glass material.

Prior art: none of the documents of the search report suggests a probe having an end being encased within a glass material.

indicator, being changed either when a drop in performance is seen or using a calculated figure based on usage time under particular conditions. This can lead to early replacement of the bubbler when potentially another 10% of material in the bubbler could be used.

Clearly it would be desirable to provide an accurate probe for monitoring the level of organometallic precursor contained within a bubbler. However, problems arise with the provision of a suitable probe that has the necessary robustness and compatibility with the organometallic compounds with which it comes into contact. In this respect, the probe must not react with the chemicals contained within the vessel to ensure high purity metal deposits are achieved. Additionally, it should not impair the airtight seal of the bubbler since the contents are air-sensitive and the pressure within the vessel should be maintained.

It is an object of the present invention to provide a method of continuously monitoring the level of a reactive liquid in a vessel, particularly but not exclusively the level of a metalorganic compound, which aims to overcome, or at least alleviate, the abovementioned drawbacks.

A further object of the present invention is to provide an apparatus for continuously monitoring the level of a liquid in a vessel, particularly but not exclusively an metalorganic compound, that aims to overcome, or at least alleviate, the abovementioned drawbacks.

Accordingly a first aspect of the present invention provides a method for monitoring the level of an organo metallic compound in a vessel, the method comprising the steps of inserting at least one metallic probe into a vessel to act as a first electrode, one end of the probe being encased in a glass material, hermetically

sealing the end of the probe encased in a glass material within the vessel, providing a further electrode spaced apart from the first electrode to form a capacitor, applying an electric current to the capacitor and monitoring the capacitance thereof.

A second aspect of the present invention provides an apparatus for monitoring the level of a liquid in a vessel, the apparatus comprising at least one metallic probe hermetically sealed within the vessel to act as a first electrode, the sealing end of the probe being encased within a glass material, a second electrode spaced apart from said first electrode to form a capacitor, means for applying an electric current to the capacitor and means for monitoring the capacitance thereof.

It is to be appreciated that the two spaced apart electrodes are provided in order to set up the necessary dielectric therebetween. The capacitance of the capacitor thus formed will vary with the amount of liquid between the probes thereby enabling the level of liquid within the vessel to be monitored.

Preferably the vessel itself is a metallic container, for example being of stainless steel, and acts as the second electrode. Alternatively, two parallel probes could be sealed within the apparatus to act as the first and second electrodes respectively.

The metallic probe that is inserted into the vessel may be in the form of a rod, a flat elongated plate or tube. The probe may be hollow or solid. Preferably the probe is made of stainless steel.

The probe according to the present invention is particularly suitable for use in monitoring levels of metalorganic compounds.

The probe is preferably attached to a port at the top of a vessel, the vessel generally being in the form of a bubbler, that contains an inlet and an outlet pipe. One

end of the probe is encased within a glass material and this is hermetically sealed within the port. More preferably, the probe is sealed within a mounting or cap that is inserted into the port of the vessel. Preferably, the mounting is provided with electrical connections for the probe, for example in the form of a bayonet type connector, such as a BNC connector.

A coating, for example of an elastomeric material such as Teflon™, may be applied over at least a part of the probe that extends from the seal.

More preferably, at least the part of the probe that is encased in the glass material to form a metal to glass seal comprises a nickel alloy, more preferably being typically 70% nickel. More preferably, the alloy is made precipitation hardenable by the additions of aluminium and/or titanium thereto. Suitable alloys include those sold under the trade names Inconel X-750, Inconel 600 or Kovar. More preferably, an Inconel X-750 alloy is used. The glass that is sealed around this part of the probe is preferably a borosilicate glass. It is preferable for the probe to be sealed to the glass by such a matched seal.

The glass material may then be sealed within the mounting. The mounting is preferably comprised of a nickel alloy, such as Inconel X-750. The mounting preferably incorporates a gasket face seal fitting, such as a VCR profile at a connecting face and internal faces to suit the fit of an electrical connector and the glass-to-metal hermetic seal. The gasket is preferably one which is deformed on tightening to provide a secure metal-metal seal. The electrical connector is preferably surrounded by a layer of insulating material, such as a polyether ether ketone (PEEK).

Conventional means may be provided for applying an AC or DC source to the probe, together with monitoring means, such as a capacitance meter, for measuring a

change in capacitance. Preferably, a recorder is also provided for recording the change in capacitance. The recorder may include display means, such as a liquid crystal display. It is preferable for the capacitance to be continuously monitored thereby providing a continuous reading of the level of liquid in the vessel.

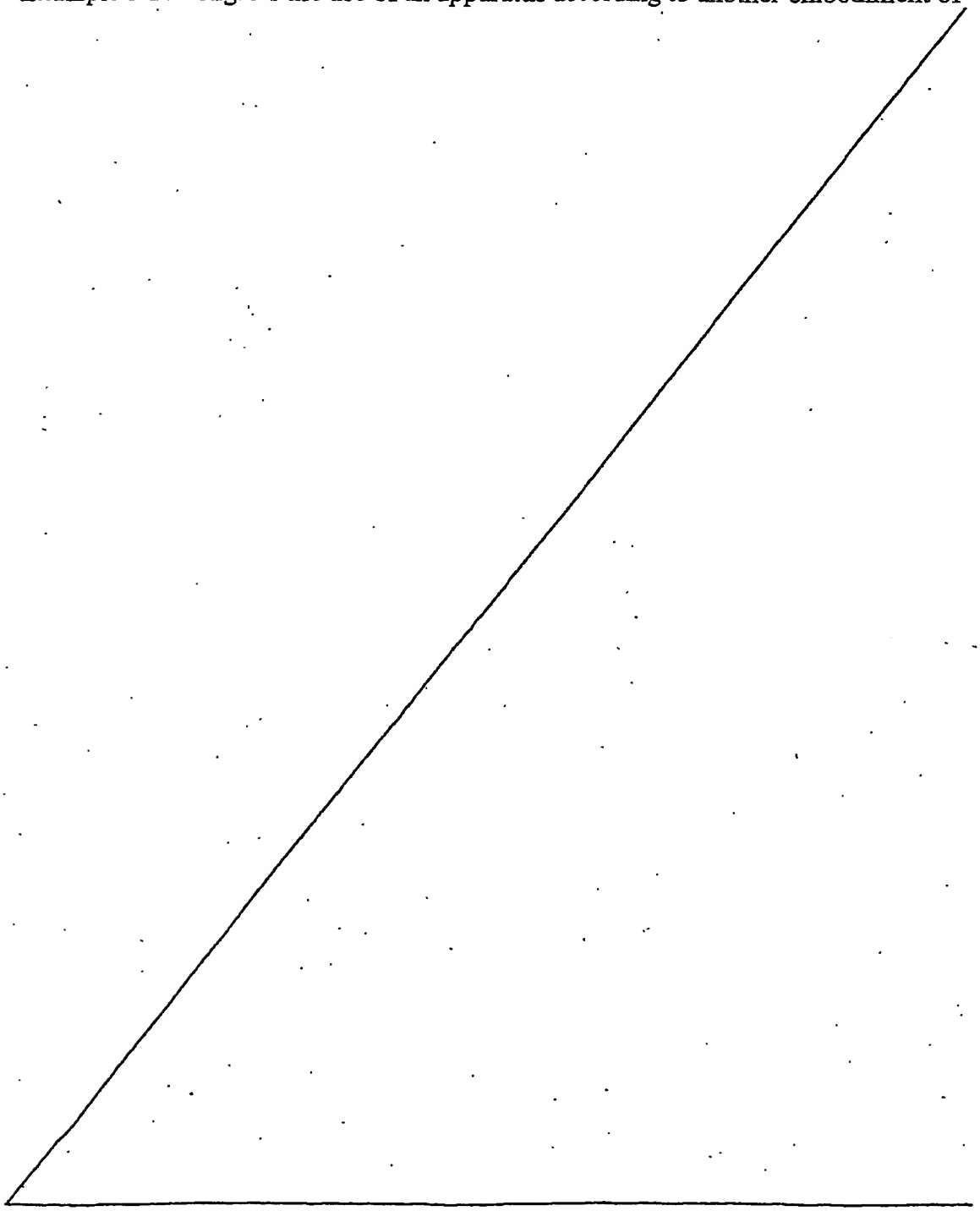
Preferably, the apparatus includes means for calibration of the system whereby a particular capacitance corresponds to a particular volume of liquid within the vessel. For example, the recording means could be set at a value of "0" for a capacitance recorded for an empty vessel and could be set "100" for the capacitance recorded for a full vessel. Preferably, the apparatus is calibrated to respond to particular characteristics of the liquid contained within the vessel. Additionally, the apparatus may be adapted to provide the rate of removal or addition of the liquid to the vessel.

In a preferred embodiment of the present invention there is provided a bubbler containing an metalorganic compound, the bubbler comprising a sealed metallic container having an inlet pipe, an outlet pipe and a dip-tube and further comprising a metallic probe hermetically sealed within the container, the sealing end of the probe being encased in a glass material, the container and the probe forming a capacitor, means for applying an electric current to the capacitor and monitoring means for measuring the capacitance thereof.

It is to be appreciated that the metallic probe may be hermetically sealed within the bubbler as hereinbefore described.

For a better understanding of the present invention and to show more clearly how it may be carried into effect reference will now be made to the following Examples in which Example 1 investigates the use of an apparatus according to one embodiment of the present invention in measuring the changing level of

trimethylgallium (TMG) in a bubbler, Example 2 investigates the use of the same apparatus in measuring the changing level of trimethylaluminium (TMA) in a bubbler, Example 3 investigates the use of an apparatus according to another embodiment of



CLAIMS

1. An apparatus for monitoring the level of a liquid in a vessel (2), the apparatus comprising at least one metallic probe (10) hermetically sealed within the vessel to act as a first electrode, the sealing end (22) of the probe being encased within a glass material, a second electrode spaced apart from said first electrode to form a capacitor, means (14) for applying an electric current to the capacitor and means for monitoring the capacitance thereof.

2. An apparatus as claimed in claim 1 wherein the vessel itself is a metallic container and acts as the second electrode.

3. An apparatus as claimed in claim 1 or claim 2 wherein the probe is made of stainless steel.

4. An apparatus as claimed in any one of claims 1 to 3 wherein the glass material is a borosilicate glass.

5. An apparatus as claimed in any one of claims 1 to 4 wherein the probe is hermetically sealed within a port provided at the top of the vessel.

6. An apparatus as claimed in claim 5 wherein the probe is sealed within a mounting or cap (30) that is inserted into the port of the vessel.

7. An apparatus as claimed in claim 6 wherein the mounting is provided with electrical connections for the probe.

8. An apparatus as claimed in any one of the preceding claims wherein the probe is provided with a coating of an elastomeric material over at least the part that extends from the seal.

9. An apparatus as claimed in any one of the preceding claims wherein at least the part (22) of the probe that is encased within a glass material comprises a nickel alloy.

10. An apparatus as claimed in claim 9 wherein the alloy is an Inconel or Kovar alloy.

11. An apparatus as claimed in claim 9 or claim 10 wherein the alloy contains aluminium and/or titanium.

12. An apparatus as claimed in claim 11 wherein the alloy is Inconel X-750.

13. An apparatus as claimed in any one of claims 6 to 12 wherein the mounting (32) is made of a nickel alloy.

14. An apparatus as claimed in claim 13 wherein the alloy is Inconel X-750.

15. An apparatus as claimed in any one of the preceding claims further comprising monitoring means for measuring a change in capacitance.

16. An apparatus as claimed in claim 15 further comprising a recorder for recording a change in capacitance.

17. An apparatus as claimed in claim 16 further comprising display means for displaying the level of liquid in the vessel.

18. An apparatus as claimed in any one of the preceding claims further comprising calibration means for calibration of the apparatus whereby a particular capacitance corresponds to a particular volume of liquid within the vessel.

19. The use of an apparatus as claimed in any one of the preceding claims in monitoring the level of organometallic compounds.

20. An apparatus as claimed in one of claims 1 to 18 wherein the vessel is a bubbler.

21. A bubbler containing an organometallic compound, the bubbler comprising a sealed metallic container having an inlet pipe (4), and outlet pipe (8) and a dip tube (6) and further comprising a metallic probe (10) hermetically sealed within the container, the sealing end (22) of the probe being encased within a glass material, the container and the probe forming a capacitor, means for applying an electric current to the capacitor and monitoring means for measuring the capacitance thereof.

22. A method for monitoring the level of an organometallic compound in a vessel, the method comprising the steps of inserting at least one metallic probe (10) into a vessel (2) to act as a first electrode, one end (22) of the probe being encased in a glass material, hermetically sealing the end of the probe encased in a glass material within the vessel, providing a further electrode to form a capacitor, applying an electric current to the capacitor and monitoring the capacitance thereof.